Hardness Measurement and Evaluation of Double-layer Films on Material Surface

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Abstract: A method for hardness measurement and evaluation of double-layer thin films on the material surface is proposed. Firstly, it is studied how to obtain the force-indentation response with the finite element method when the indentation is less than 100 nanometers, in which current nanoindentation experiments have no reliable accuracy. The whole hardness-displacement curve and fitted equation are obtained. At last, a formula to predict the hardness of the thin film on the material surface is derived and favorably compared with experiments.

Key words: double-layer films; hardness; nanoindentation; finite element simulation

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摘 要: 给出了基于纳米硬度试验的二层薄膜的硬度测算方法。首先利用有限元方法探讨了二层薄膜- 基体材料系统的硬度随压痕深度变化的规律,进而推导和验证了材料系统硬度- 位移曲线的拟合公式,最后给出了以压痕实验为基础的二层薄膜材料硬度的计算公式。

关键词: 两层薄膜; 硬度; 纳米压痕; 有限元模拟

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With the rapid development of surface engineering technologies and their successful applications in various sectors of industry, the micro-scale indentation test, especially the nanoindentation test has become one of the basic methods to probe the mechanical properties of a lot of new engineering films. However, for films from one to hundreds of nanometers thick, the difficulties associated with the measurements of thin film properties arise from the effects of the substrate. In fact, the hardness obtained from experiment shows the combined action of thin films and substrate. Therefore, how to evaluate the hardness of thin films becomes an important problem to be overcome urgently.

A lot of literature[1-4] has investigated the measurement methods of thin film hardness. In general, the one-tenth rule is a fundamental rule to evaluate the hardness of thin films, *i.e.*, when the indentation depth is less than 10% of the film thickness, the contribution of the substrate on the experimental result will be less than 2%. According to this rule, when the film thickness is less than 200nm, the indentation depth should be less than 20nm in order to obtain intrinsic film hardness, which will be beyond the ability of the present indentation technique[5].

In the author's former work[6], for a single-film/substrate composite, the formula

$$H_f = H_s + (H - H_s)\exp(2.2h/t) \qquad (1)$$

was proposed to predict film hardness according to the experimental results. H_f and H_s are the hard-

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ness of the film and substrate, respectively. t is the thickness of the film, and H is a measured hardness while the depth of indentation is h.

The film hardness of multi-film/substrate systems can hardly be directly determined through indentations because of their complex constitutions, especially when the film is between the first-layer film and substrate. Though much work has been performed, there is still no efficient and effective means to solve this problem.

In the present paper, the hardness regularities of double-film/substrate systems are investigated through combining the finite element method (FEM) with nanoindentation experiments. The geometrical shape of the nanoindenter is precisely simulated and the loading-unloading process of indentation is represented with a three-dimensional FEM.

1 Simulation of Nanoindentation

1.1 Geometrical shape and mesh

As shown in Fig. 1, Berkovich indenter, the standard indenter of nanoindentation equipment, has the geometrical shape of a triple pyramid. According to symmetry, 1/6 of the indenter and composite is considered. The double-film/substrate system is simulated as three parts: first-layer, middle-layer films and substrate. Fig. 2 shows the FEM mesh. The three-dimensional model was described in detail in Ref. [6]. The indenter, made in diamond, should be handled as an elastic body with the Young's modulus 1141GPa and Poisson's ratio 0.007. The material properties of the specimen are shown in Fig. 3, in which E is Young's modulus, Y is yield strength, and E_T denotes hardening modulus.

1.2 Characteristics of FEM results

Fig. 4 shows the hardness-displacement curves obtained by FEM from a double-film/substrate system. In this model, the hardness values of the first-layer, middle-layer films and substrate are in a descending order. To investigate the relationship between composite hardness and that of films and substrate, material models of the composite, films

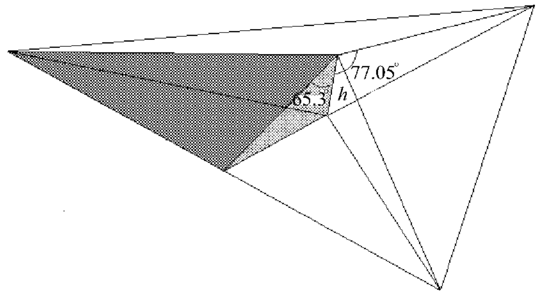


Fig.1 Shape and parameters of Berkovich indenter

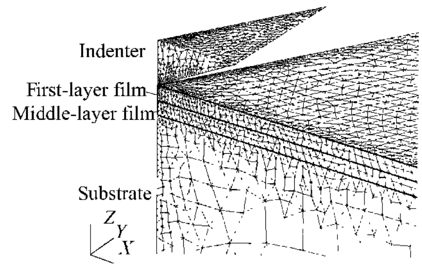


Fig.2 Finite element model of double-film/substrate

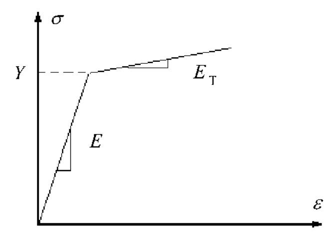


Fig.3 Stress-strain curve of material

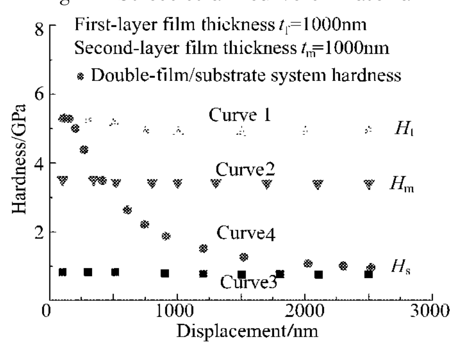


Fig.4 The hardness-displacement curves obtained with FEM

and substrate are independently established to calculate their hardness, respectively. Fig.5 gives four hardness-displacement curves, in which curves 1, 2 and 3 represent the first-layer, middle-layer films and substrate, respectively. From Fig. 4, it can be seen that the curves 1, 2 and 3 vary flatly and straightly, which is in accordance with the previous researc[7] of pure substance hardness. Curve 4 represents the composite that locates between curve 1 and curve 3, and it matches curve 1 for small indentation because the hardness is mainly determined by films rather than by the substrate.

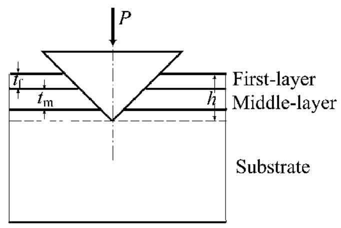


Fig. 5 The sketch map of double-film/substrate system and nanoindenter

2 Regularity of Hardness Variation with Indentation Depth

2.1 Deduced formula of hardness-displacement curves

In Ref. [6], the hardness fitted equation of the single-film/substrate system was given as

$$H = H_s + (H_f - H_s)\exp(-\gamma h/t) \quad (2)$$

where γ is a parameter to be determined from experiment and FEM results. Eq. (2) indicates the combined actions of thin films and substrate on the measured hardness. It can be factored as

$$H = H_s - H_s\exp(-\gamma h/t) + H_f\exp(-\gamma h/t) \quad (3)$$

where $\exp(-\gamma h/t)$ denotes the influence of the material from surface to t-depth on the measured hardness. So $H_s - H_s\exp(-\gamma h/t)$ and $H_f\exp(-\gamma h/t)$ represent the influence of the substrate and film on hardness, respectively.

The hardness of a double-film/substrate system reflects the combined action of the first-layer, middle-layer films and substrate. Therefore, the same method as for single-film/substrate systems is adopted to analyze the regularity of hardness (H) and indentation depth(h). Then the fitted equation is assumed as

$$H = H_s - H_s\exp\left(\frac{-\gamma h}{t_f + t_m}\right) + H_m\exp\left(\frac{-\gamma h}{t_f + t_m}\right) - H_m\exp\left(\frac{-\gamma h}{t_f}\right) + H_f\exp\left(\frac{-\gamma h}{t_f}\right) \quad (4)$$

where H_f, H_m and H_s are the hardness of the first-layer, middle-layer films and substrate, and t_f and t_m are thickness of the first-layer and middle-layer films, respectively. γ is a parameter to be determined, and H is the hardness of the composite while the indentation depth is h. In Eq. (4),

$$H_s - H_s\exp\left(\frac{-\gamma h}{t_f + t_m}\right),$$

$$H_m\exp\left(\frac{-\gamma h}{t_f + t_m}\right) - H_m\exp\left(\frac{-\gamma h}{t_f}\right) \text{ and}$$

$H_f\exp(-\gamma h/t_f)$ denote the influence of the substrate, middle-layer and first-layer films on the measured hardness(H), respectively. Then Eq. (4) can be written as

$$H = H_s + (H_m - H_s)\exp\left(\frac{-\gamma h}{t_f + t_m}\right) + (H_f - H_m)\exp\left(\frac{-\gamma h}{t_f}\right) \quad (5)$$

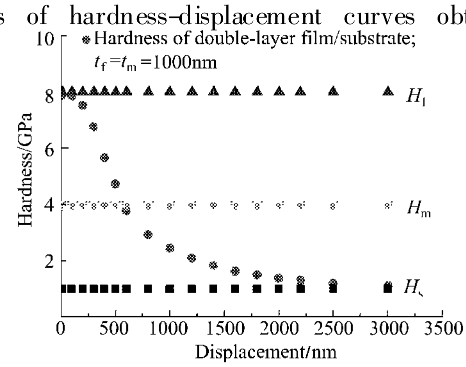
2.2 Six types of hardness-displacement curves

While the hardness of the first-layer, middle-layer films and substrate is not equal to each other, the hardness-displacement curves of the composites can be classified as six types according to the magnitude order of H_f, H_m and H_s, as shown in Figs. 6. It can be seen that if H_f, H_m and H_s are in an ascending ($H_f > H_m > H_s$) or a descending order $H_f < H_m < H_s$, the hardness-displacement curves vary correspondingly, as shown in Fig. 6(a) and Fig. 6(b). Otherwise, the shapes of curves are shown in Fig. 6s(c), (d), (e) and (f).

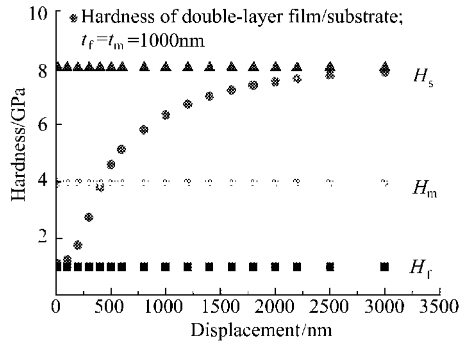
3 Evaluation of Thin Film Hardness

3.1 Verification of fitted formula

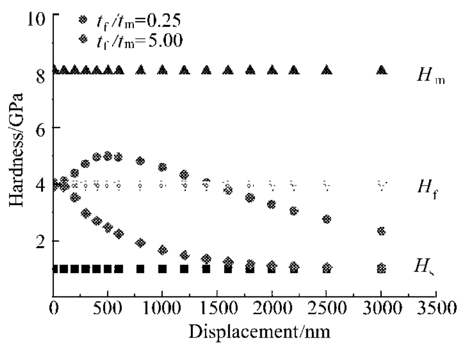
Eq. (5) is a deduced formula to relate the hardness of the composite to that of its components, with a parameter(γ) to be determined. In the present work, Eq. (5) is tested by fitting six types of hardness-displacement curves obtained



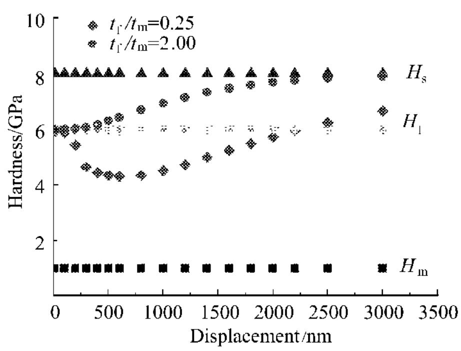
(a) $H_f > H_m > H_s$



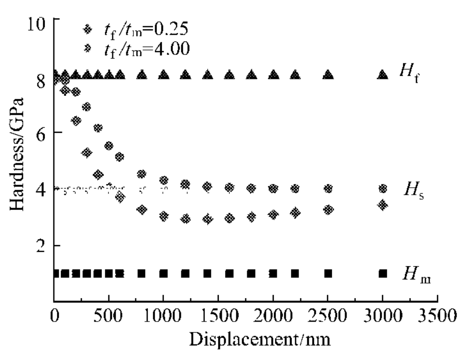
(b) $H_f < H_m < H_s$



(c) $H_m > H_f > H_s$



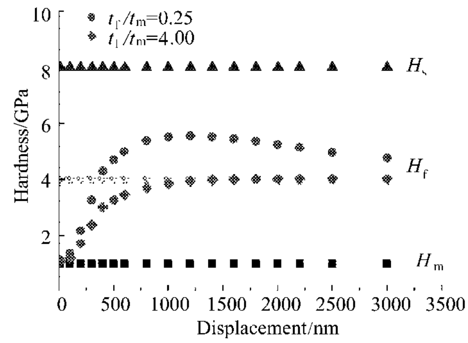
(d) $H_s > H_f > H_m$



(e) $H_f > H_s > H_m$

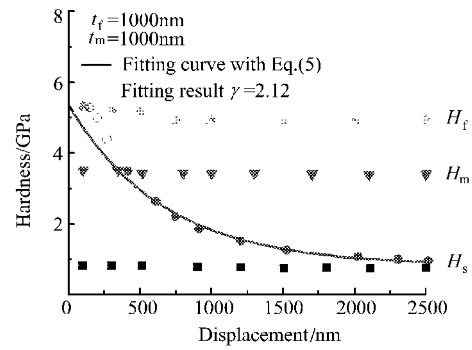
through parameter combinations of films and substrate which are shown in Table 1, and γ is determined.

Fig. 7 (a) and Fig7. (b) show the fitted results of two types of hardness-displacement curves. In addition, all types of curves were fitted and the results showed favorable. To be clear, on-

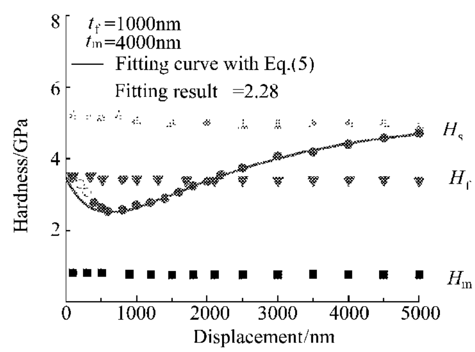


(f) $H_m > H_s > H_f$

Fig. 6 The shape of hardness-displacement curve



(a) $H_f > H_m > H_s$



(b) $H_s > H_f > H_m$

Fig. 7 The fitting curve

ly two results are given in Fig. 7. In order to compare, the authors also give the hardness of the first-layer, middle-layer and substrate. Eq. (5) is applied to fit the data with more than 200nm-depth, because in this range the hardness can be obtained accurately by nanoindentaion experiments. Much fitted results indicate that γ is in the range of 2. 2±0. 2. In Eq. (5) , let γ= 2. 2. Then

$$H = H_s + (H_m - H_s)\exp\left(\frac{-2.2h}{t_f + t_m}\right) +$$

$$(H_f - H_m)\exp\left(\frac{-2.2h}{t_f}\right) \tag{6}$$

Eq. (6) is the fitted formula of hardness-displacement curves, which denotes the hardness relationship between the composite and its components.

Table 1 Material parameters used in models of double-film/substrate system

Yield strengths Y(MPa)	10	50	100	400	800	1600	3200
Yang's moduli E(GPa)	26	64	256	384	640		
Hardening moduli E_T(GPa)	0.1	0.5	1.0	5.0	10.0	20.0	

(Poisson's ratio $v = 0.28$)

3.2 Application of fitted formula

For a double-film/substrate system, Eq. (6) denotes the relationship between the composite, films and substrate. It also reflects the influence of film thickness on hardness(H). In Eq. (6), let

$$\alpha = \exp\left(\frac{-2.2h}{t_f + t_m}\right), \beta = \exp\left(\frac{-2.2h}{t_f}\right)$$

Then the formula to predict the hardness of the first-layer film is

$$H_f = H_m + [H - H_s - (H_m - H_s)\alpha]/\beta \tag{7}$$

and the formula for the middle-layer film is

$$H_m = \frac{H - H_s + H_s\alpha - H_f\beta}{\alpha - \beta} \tag{8}$$

Eqs. (7) and (8) have the application qualifications as follows:

(1) The substrate hardness H_s, film thickness t_f and t_m must be known or can be measured.

(2) The composite hardness H is measured with the indentation depth satisfying $0.2(t_f + t_m) < h < 1.2(t_f + t_m)$.

(3) One of the hardness values H_f and H_m had better be known, or their relationship is certain, such as $H_f = g(H_m)$. If they are all unknown, at least two hardness values H_1 and H_2 of the composite in different depths should be measured to obtain H_f and H_m from Eq. (6).

4 Conclusions

(1) According to the regularity of single-film/substrate systems, the fitted Eq. (6) of hardness-displacement curves of double-film/substrate can be deduced, which denotes the combined action of thin films and substrate on hardness.

(2) The deduced fitted Eq. (6) can be tested and determined by numerical simulation of the indentation process. Based on nanoindentation results, the hardness of thin films on the substrate can be effectively obtained from Eqs. (7) and (8).

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